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TITLE NONDESTRUCTIVE ASSAY OF PLUTONIUM RESIDUE IN HORIZONTAL STORAGE TANKS

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## NONDESTRUCTIVE ASSAY OF PLUTONIUM RESIDUE IN HORIZONTAL STORAGE TANKS\*

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### ABSTRACT

Aqueous plutonium recovery and purification processes often involve the temporary storage of plutonium solutions in holding tanks. Because plutonium is known to precipitate from aqueous solutions under certain conditions, there is a continuing need to assay emptied tanks for plutonium residue. A portable gamma spectrometer system, specifically designed for this purpose, provides rapid assay of such plutonium residues in horizontal storage tanks. A means is thus available for the nondestructive analysis of these tanks on a regular schedule to ensure that significant deposits of plutonium are not allowed to accumulate.

### INTRODUCTION

The Los Alamos Plutonium Facility at TA-55 uses a nitrate anion exchange process (Figure 1) to recover and purify plutonium from a wide variety of materials. The major process steps are (1) dissolution of the impure materials in fluoride-catalyzed nitric acid, (2) chemical treatment of this solution to ensure high retention of plutonium on the anion exchange column, (3) sorption of plutonium on the column while impurities wash through, (4) elution of purified plutonium as Pu(III), (5) precipitation of the eluted plutonium as an oxalate salt, and (6) calcination of plutonium oxalate to plutonium dioxide.

Several stages of this process require the temporary storage of plutonium-containing solutions in horizontal storage tanks of cylindrical, always-safe geometry (Figure 2). These solutions, under certain conditions, have been known to deposit a portion of the plutonium as an insoluble residue. It is essential that any such plutonium residues are promptly detected and recovered for reasons that include both accountability and criticality concerns. A need therefore exists for a nondestructive assay technique that can be used to monitor these storage tanks on a regular basis.

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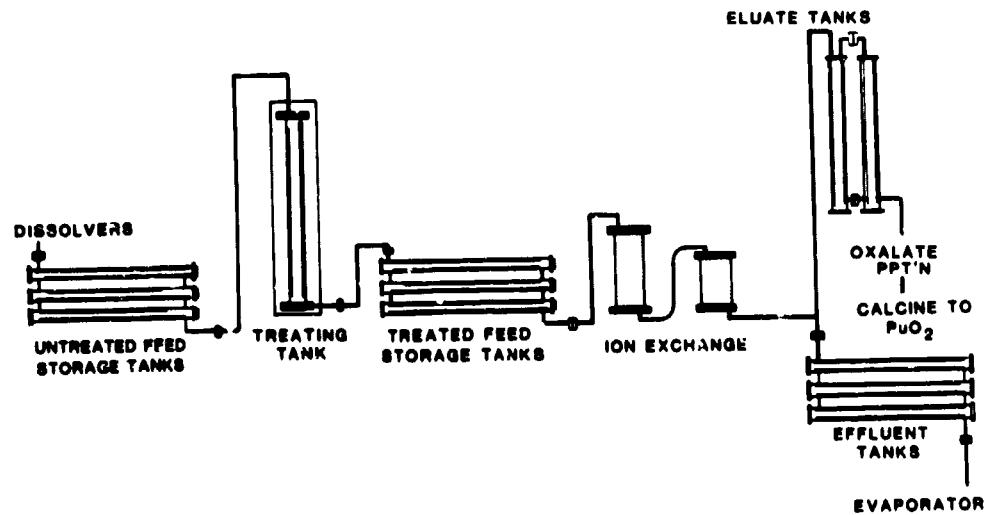


Figure 1. Schematic of the Nitrate Anion Exchange Process used at Los Alamos

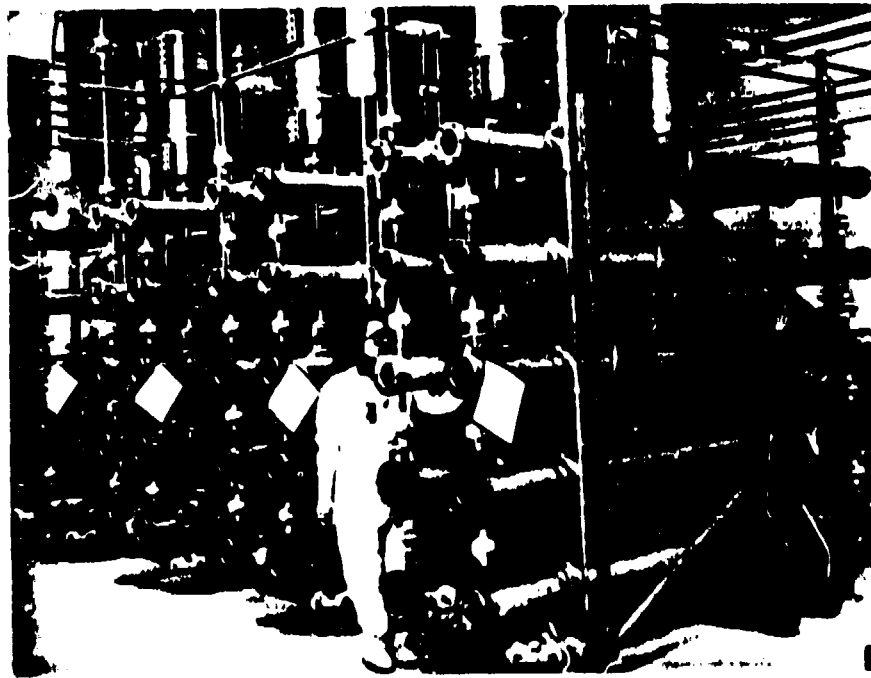


Figure 2. Horizontal Storage Tanks for Plutonium Solutions

## GAMMA ASSAY SYSTEM

A portable gamma spectrometer system was assembled for measuring plutonium residue in horizontal storage tanks. The gamma detector is a 2" by 2" NaI(Tl) scintillation crystal coupled to an integral-design photomultiplier tube. The detector unit is enclosed in a cadmium-lined lead shield (Figure 3), whose base fits the curvature of the tanks to be assayed. A sliding lead shutter between the detector and the tank provides a choice between a collimated beam hole, or a solid 1/2" thickness of lead shielding. The collimated beam that reaches the detector is further attenuated by a 0.030" tungsten absorber that eliminates much of the low-energy gamma radiation of americium and plutonium. Gamma radiation from adjacent, lower tanks in the "line-of-sight" of the detector is reduced by covering these tanks with a 1/4" thickness of lead sheet (Figure 4). The amplified signal is processed by a 2048-channel multichannel analyzer on a wheeled cart to facilitate system mobility (Figure 4).



Figure 3. Gamma Detector/Shield Assembly

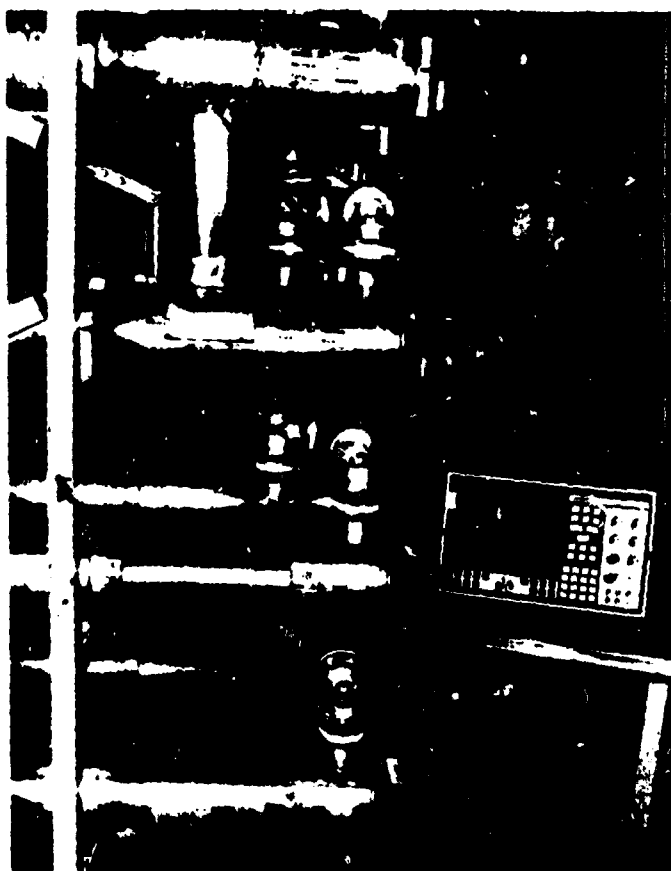


Figure 4. Nondestructive Gamma Assay System in Use

#### OPERATION

Each storage tank, after draining the solution to eliminate the contribution of dissolved plutonium, is measured at predetermined positions along the length of the tank. We typically measure at the midpoint, and at two-foot increments from the midpoint toward both ends, which corresponds to five measurements for a ten-foot tank. Two 100-second counts are taken at each measurement position, one with the shutter open, and one with the shutter closed. The area under the 414-keV gamma peak of Pu-239 is integrated in each case. The difference between these two integrated areas is the background-corrected count for that position. (Background correction is essential because the surrounding tanks can contain kilogram quantities of plutonium in solution.) The sum of the five individual background-corrected counts represents the total plutonium residue in the tank.

Note: The decision to count only five equally spaced positions on a ten-foot tank was an arbitrary compromise between assay time and precision. Better precision would result from counting the tank at more positions that are separated by smaller distances, but at a cost of

increasing the assay time. Likewise, extending the counting time beyond 100 seconds would improve the measurement precision by increasing the assay time.

#### CALIBRATION

The most reliable way to calibrate any radioassay system is to use calibrations standards that simulate as closely as possible the actual objects that will be assayed. We have carried this philosophy to the limit by using the storage tanks themselves as calibration standards. This is done by assaying a tank immediately before and immediately after the plutonium in that tank is removed. The difference between the before- and after-cleaning assay values is directly attributable to the amount of plutonium removed in the cleanout operation. Thus, a direct correlation between plutonium content and assay count-rate is obtained. (Plutonium removed in these cleanout solutions is always measured by an independent and highly reliable analytical technique.)

Each tank cleaned in this manner provides an independent calibration factor. These individual calibration factors will, of course, differ from each other as a result of the nonuniform deposition of plutonium in the tanks, and other factors that contribute to measurement variability. As additional tanks are cleaned, additional calibration factors are generated. From these multiple calibration factors, a calibration curve may be calculated. This curve reflects the combined effects of plutonium deposit thickness variation and all other system variables. Furthermore, this curve is continuously updated and refined by the inclusion of additional calibration data to provide the best basis for assaying an "unknown" tank. The standard deviation of the fit of the individual factors to a least squares line provides a realistic estimate of the uncertainty associated with a single assay.

A check-source of approximately 200 milligrams of plutonium was prepared in a double-welded capsule that fits snugly within the collimator hole of the lead shutter. This check-source is counted each day before the assay system is used to ensure that the entire system is operating properly. Minor adjustments for gain shift are made when necessary. Major differences from previous count-rates, however, indicate that the assay measurements are unreliable. In this case, the system may be in need of a major adjustment or repair.

#### RESULTS

The gamma assay system is intended to provide a reliable estimate, rather than a quantitative measurement of deposited plutonium. The measurement of nonuniform deposits of plutonium along the length of a ten-foot tank that is surrounded by other tanks that contain kilogram amounts of plutonium, is hardly an ideal situation for making quantitative measurements. Nevertheless, the precision of these assays, based on a calibration curve generated from six individual calibration factors (Figure 5), was better than expected. The rel-

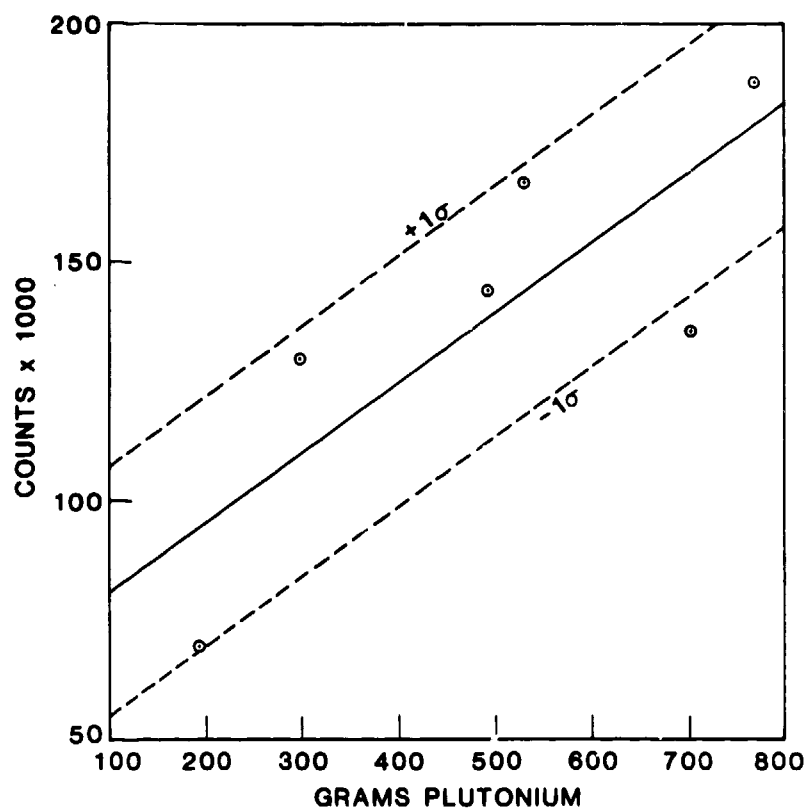


Figure 5. Experimental Calibration Curve  
for Plutonium Residue Assay

ative standard deviation for measuring the plutonium residue in a single tank ranged from 30% for 200 grams of plutonium, to 13% for 800 grams of plutonium. These precision estimates are expected to improve as additional calibration factors are determined and incorporated into the curve.